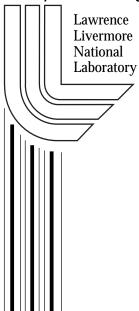
Carbon Nanotube Based Microfluidic Elements for Filtration and Concentration

O. Bakajin, N. Ben-Barak, J. Peng, A. Noy

This article was submitted to Micro Total Analysis Systems Squaw Valley, CA October 5-9, 2003

June 25, 2003





DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

This report has been reproduced directly from the best available copy.

Available electronically at http://www.doc.gov/bridge

Available for a processing fee to U.S. Department of Energy
And its contractors in paper from
U.S. Department of Energy
Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831-0062
Telephone: (865) 576-8401

Facsimile: (865) 576-5728 E-mail: reports@adonis.osti.gov

Available for the sale to the public from U.S. Department of Commerce National Technical Information Service 5285 Port Royal Road Springfield, VA 22161 Telephone: (800) 553-6847

Facsimile: (703) 605-6900 E-mail: orders@ntis.fedworld.gov

Online ordering: http://www.ntis.gov/ordering.htm

OR

Lawrence Livermore National Laboratory Technical Information Department's Digital Library http://www.llnl.gov/tid/Library.html

CARBON NANOTUBE BASED MICROFLUIDIC ELEMENTS FOR FILTRATION AND CONCENTRATION

Olgica Bakajin, Nadav Ben Barak, Jenet Peng, Aleksandr Noy

Biosecurity Nanoscience Laboratory, Chemistry & Materials Science Directorate, Lawrence Livermore National Laboratory, Livermore, CA

ABSTRACT

We have developed a method for integration of patterned arrays of carbon nanotubes or the "nanotube mesh" into microfabricated channels. The method includes standard lithographic methods for patterning and etching the substrate, followed by catalyst patterning, CVD deposition of nanotubes, and anodic bonding of coverslip top. We will describe a carbon nanotube filtering device fabricated using this method and discuss the use of carbon nanotube arrays as molecular concentration and separation media.

KEYWORDS

filtration, concentration

INRODUCTION

Filtration of the sample prior to processing is an essential step when performing analysis in microfluidic devices. Since microfluidic channels are small, particle contamination can cause operational problems. On chip filtration methods are desirable because they allow handling of small amounts of liquid, which could not be processed via conventional filtration methods and are sufficient for analytical on-chip processing. Concentration of the analyte is another essential step that increases detection efficiency. Carbon nanotube microfluidic elements have a potential for both filtration and concentration.

A common approach for microfluidic filtering has been creation of pillar structures or flow restrictions [1, 2]. The limitation of these methods is the resolution of the patterning technique employed. Filtration of sub-micron particles using lithographically patterned structures would require expensive fabrication methods such as e-beam lithography. Alternatively, emulsion photopolymerization has been used to create filters of various pore sizes but this method also creates pores on the order of a micron[3]. Nano-scale filters with pore sizes as small as 10nm have been made using sacrificial layer technology [4].

Carbon nanotubes, with their unique properties, dimensions and a huge surface to volume ratio, have a great potential as a filtration, separation and concentration medium for various chromatographic applications. Carbon nanotube mesh self assembles into a robust nano-porous structure, which eliminates the need for nano-lithographically patterned features. The mesh has pores on the order of 10-50nm, the surface-to-volume ratio superior to the packed bead columns and the surface that can be modified with polymers that incorporate particular chemical functionality [5]. A possibility of patterning carbon nanotube arrays in specific parts of a microfabricated device allows easy integration into microfluidic devices and gives this approach a great advantage over more conventional separation media that requires column packing such as silica beads. Nanotube arrays can also be tuned to a particular application since it is possible to control nanotube size, density and orientation in the growth process.

METHODS

We use standard fabrication techniques to pattern a 5 nm thick film of iron catalyst in the desired area of the channel etched in silicon. Afterwards we used CVD growth technique to grow carbon nanotube arrays, ranging from 5 μm to 100 μm in height. The catalyst is first oxidized to form islands on which individual nanotubes start to grow. We then use pyrolysis of a mixture of ethylene, hydrogen and argon at 850C which results in a mesh of nanotubes. After we grow the mesh, we seal the devices by anodically bonding a top layer onto them in vacuum. For filtration purposes, we grow the mesh of nanotubes so that it is a few microns taller than the depth of the channel. We rely on good elastic properties of the nanotube mesh and pack all of the tubes into the channel in order to assure that there is no gap between the glass and the nanotubes. Figure 1 shows carbon nanotubes grown in a silicon channel.

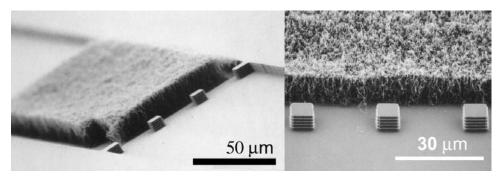


Figure 1: SEM images of the carbon nanotube mesh inside the microfabricated channels in silicon.

RESULTS

In order to demonstrate filtering capability of nanotube elements we used a mixture of fluorescent beads that was moved through the channel using pressure driven flow. Figure 2 shows the fluorescent beads of 198nm diameter concentrated at the carbon nanotube filter. Beads were successfully released from the filter by applying the back-pressure. The solutions contained TWEEN-20 (Sigma) in order to render carbon nanotube surface hydrophilic and facilitate introduction of water into the filter through capillary action. With filter lengths of several hundreds of microns, pressures on the order of a few to a few tens of psi were sufficient to drive the fluid. Since nanotube patches that are only a few tens of microns long should be sufficient for filtering, we do not anticipate the filter impedance to be a practical difficulty. Studies are currently performed to quantitatively characterize the hydrodynamic resistance of the filters as well as to determine the minimum size of the particle that can be filtered in our devices.

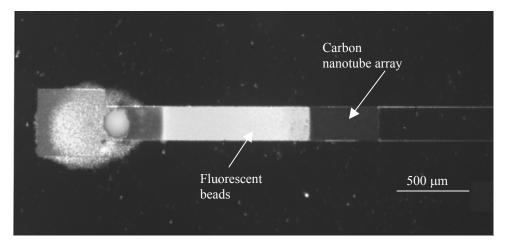


Figure 2 Fluorescent beads (D=198nm) concentrated at the carbon nanotube filter. This channel is 6.5 µm deep. Fluid flow was left to right.

DISCUSSION AND CONCLUSION

We have demonstrated the filtering capability of the carbon nanotube arrays and demonstrated a simple method for integration of sub-micron filters into microfluidic devices. In addition, we are exploring use of these devices and of microfluidic channels with carbon nanotube coating as pre-concentrators for micro-gas chromatography and as

liquid chromatography columns. We are also performing adhesion measurements between various chemical functionalities and the curved graphite-like nanotube surface using chemical force microscopy. A combination of these measurements with *ab-initio* modeling will allow us to understand the molecular interactions with the nanotube surfaces and apply rational design to nanotube based chromatography systems.

ACKNOWLEDGEMENTS

This work was performed under the auspices of the US Department of Energy by UC LLNL under contract #W-7405-Eng-48 with funding from the LDRD program.

REFERENCES

- [1] Andersson, H., W. van der Wijngaart, and G. Stemme, *Micromachined filter-chamber array with passive valves for biochemical assays on beads*. Electrophoresis, 2001. **22**(2): p. 249-257.
- [2] He, B., L. Tan, and F. Regnier, *Microfabricated filters for microfluidic analytical systems*. Analytical Chemistry, 1999. **71**(7): p. 1464-1468.
- [3] Moorthy, J. and D.J. Beebe, *In situ fabricated porous filters for microsystems*. Lab on a Chip, 2003. **3**(2): p. 62-66.
- [4] Chu, W.H., et al., *Silicon membrane nanofilters from sacrificial oxide removal.* Journal of Microelectromechanical Systems, 1999. **8**(1): p. 34-42.
- [5] O'Connell, M.J., et al., *Reversible water-solubilization of single-walled carbon nanotubes by polymer wrapping.* Chemical Physics Letters, 2001. **342**(3-4): p. 265-271.